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L. R. BURDICK AND J. F. BARKLEY

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UNITED STATES DEPARTMENT OF THE INTERIOR - BUREAU OF MINES

EFFECT OF SULFUR COMPOUNDS IN THE AIR ON VARIOUS MATERIALS[1]

By L. R. Burdick[2] and J. F. Barkley[3]

This paper presents a review of published investigations dealing with the importance of sulfur compounds in the deterioration of materials or providing some basis for estimating the extent of loss chargeable to such air pollution. Materials included are cement, stone, metals, paint, leather, paper, and cloth. The investigations may be divided into two classes, one of the laboratory type in which the sulfur content and the humidity of the air were controlled and the other dealing with the exposure of materials to atmospheres not under control. The first type of work has shown that cement is subject to deterioration by the action of sulfur dioxide gas. The experiments with stone indicate that although limestone takes up sulfur dioxide from the air, moist sulfur dioxide-air mixtures alone cannot be used for establishing the comparative weathering qualities of the different types of stone by accelerated test methods. Laboratory tests indicate that sulfur compounds in the air accelerate the corrosion of iron, steel, zinc, copper, and nickel at normal air temperatures and at certain relative humidities. It has also been indicated that certain types of paint are affected by high sulfur dioxide concentrations, and that paper is attacked by sulfur dioxide in the atmosphere.

The investigations of the second type show the amount or variation of corrosion in different localities and under different types of exposure. The investigations (10) (18)[4] during which sulfur determinations were made indicate that sulfur dioxide is a main controlling factor in the corrosion of iron and steel but according to Quick (18) may not be for some nonferrous metals. In general, the laboratory tests do not give definite data on the loss due to sulfur compounds at the concentrations normally found in the air. The data from atmospheric exposures are not complete from the standpoint of the exact effect of the sulfur compounds. Damage to materials so exposed is affected by factors such as other chemical contaminants, humidity, rainfall, solid deposits, and sunlight. A simple scheme has been devised in Great Britain which is considered to provide a measure of the attack of sulfur compounds on building materials (1). Small cylinders coated

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4/ The figures in parentheses refer to items in the bibliography at the end of this paper.

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with lead peroxide paste are exposed to the atmosphere for about 1 month, after which the paste is analyzed and the results reported as milligrams of sulfur trioxide per 100 sq. cm. of cylinder per day.

REVIEW OF INVESTIGATIONS

Cement

Akiyama (2) observed that sulfur dioxide gas vigorously eroded cement sand mortar in the presence of moisture. His observation was based on laboratory work with 1.5-cm.-diameter by 3-cm. test pieces of 1:3 cement sand mortar with water-cement ratios of 0.65 and 0.75. The experiments were carried on at a temperature of 20° C., the procedure being to cure six test pieces in water for 1 week, then keep three in a moist chamber with air and three in a moist chamber with a sulfur dioxide current for periods up to 4 weeks, after which the condition of all six was noted and compared.

Stone

Investigators for the Building Research Board of Great Britain passed (3) moist air containing a constant amount of sulfur dioxide over different kinds of limestone in the laboratory and weighed the stone at intervals after suitable drying. The relative increases in weight due to sulfation were not in agreement with the observed weathering properties of the stone used in the experiments. It was concluded that factors other than the absolute degree of chemical combination with sulfur gases are of preponderating influence in determining weathering behavior.

Kaiser (4) has shown by laboratory experiment that limestone rapidly absorbs sulfur dioxide when present in high concentrations. He exposed stone samples periodically to a mixture of 80 percent air, 20 percent carbon dioxide and 1 percent sulfur dioxide (10,000 p. p. m.) in sealed containers in the presence of water vapor. He noted that a sample of gas taken from the container immediately after the above gas mixture had been let in never showed more than 0.2 percent sulfur dioxide and that no sulfur dioxide could be detected in the apparatus after a few hours.

Metals

Patterson (5) investigated the effect on pure zinc of a saturated atmosphere polluted with sulfur dioxide. He hung 2- by 2-inch test pieces by glass hooks over distilled water containing a few c. c. of 1 percent sulfurous acid. Desiccators were used and kept outdoors in the fluctuations of the day and night temperatures. The test pieces were removed at intervals of 4 to 16 days, dried, and weighed. He concluded that after the first few days corrosion attack with saturated air continues slowly when the atmosphere is clean, and more rapidly when sulfur dioxide is present as a polluting agent. The data presented show the corrosion with sulfur dioxide present to be five times more rapid, expressed in change of weight of the test piece per day. He also experimented with test pieces of mild and high-carbon steel (6) and concluded that traces of sulfur dioxide are very active in establishing the corrosion of iron under saturated conditions of the atmosphere.

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Vernon (7) exposed 5-cm. square test pieces of copper to mixtures of pure air and sulfur dioxide under a bell jar, using glass hooks for suspension. The jar was kept at a constant temperature of 77° F. At intervals of 3 to 5 days the test pieces were removed and weighed and the atmosphere within the jar was replenished. These laboratory experiments indicated that during the first 30 days of exposure the rate of corrosion of copper is much greater for atmospheres containing 0.01 percent sulfur dioxide (100 p. p. m.) than for pure air when the relative humidity is above 50 percent. His work also indicated that during this period the corrosion rate is not affected by sulfur dioxide concentrations as high as 10 percent when the relative humidity is zero and that there is a critical relative humidity in the neighborhood of 65 percent above which the rate of attack increases very rapidly with a sulfur dioxide concentration of 0.01 percent. Similar experiments with iron (8) indicated that during the first 55 days the corrosion rate with sulfur dioxide concentrations of 0.01 percent is much greater than with purified air when the relative humidity is above 70 percent. He noticed the same critical humidity of about 65 percent, above which the attack increased rapidly with 0.01 percent sulfur dioxide concentrations.

Vernon (9) also produced a creamy-white film, which he called fogging, on the polished surface of nickel by exposing the metal in the laboratory under controlled conditions to atmospheres containing 0.003 percent sulfur dioxide at 75 percent relative humidity. He concluded that the "fogging" is due partly to suspended sulfates in the air and partly (probably mainly) to the presence of small amounts of sulfur dioxide accompanied by relative humidities in excess of 70 percent, and that at lower values of relative humidity the nickel remains bright indefinitely.

Schikorr (10) exposed 45- by 30-mm. steel and cast-rion plates 3 to 4 mm. in thickness to the atmosphere on the roof of the State Board of Testing Materials, Berlin-Dahlem, and to that in Grunewald forest near Berlin for 15 months of the years 1935-36. During this time he recorded the loss of weight due to rusting, the sulfur and chlorine in the atmosphere, and the relative humidity and found that the speed of rust formation followed the seasonal variations of the sulfur value, the rusting action being decidedly more rapid in the winter when the sulfur content of the atmosphere was higher than in the summer. His data also showed highest speeds of rusting where the sulfur in the atmosphere was most concentrated, the average values for a steel being 420 grams loss in weight per square meter of area per year in Berlin-Dahlem, where the sulfur concentration as indicated by the Liesegang method (11) averaged approximately 36 mg. compared with 340 grams in Grunewald forest where the sulfur value was 8 mg., the corresponding values for cast iron being 640 grams in Berlin-Dahlem compared with 300 grams in the forest during the same period. Schikorr also conducted experiments which indicated that steels whose original rust layer had been formed in the presence of high atmospheric sulfur concentrations, rusted more rapidly when left in closed vessels in the laboratory at 93 percent relative humidity and 25° C. than those whose initial rusting had taken place under low concentrations of sulfur in the atmosphere. The results of his experimental work were such that he concluded that the main cause of atmospheric rusting is the content of moisture and sulfur compounds in the air.

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The American Society for Testing Materials through various committees has conducted several atmospheric exposure tests on metals. With the exception of those on screen wire cloth, the work did not include sulfur dioxide determinations at the test locations. These locations were chosen to represent definite atmospheric conditions, such as severe industrial (Pittsburgh, Pa.), moderate industrial (Altoona, Pa.; Rochester, N. Y.; New York, N.Y.), rural (State College, Pa., Fort Sheridan, Ill.; Phoenix, Ariz.) and sea coast (Sandy Hook, N. J.; Annapolis, Md.; La Jolla, Calif.; Key West, Fla.) Committee A-5 exposed corrugated iron and steel sheets at an angle on racks in the open air. Periodical inspections were made and corrosion failures noted, a hole or break in the sheets being called a failure. Kendall and Taylerson (12), in studying these corrosion data, have tabulated the number of failures at Pittsburgh, Fort Sheridan, and Annapolis. At Pittsburgh 90 percent of the No. 22 gage test sheets failed in 6 years exposure to the atmosphere, 58 percent in 11 years at Fort Sheridan, and 11 percent in 11 years at Annapolis. A later report (13) on the No. 22 gage sheets at Annapolis shows that 45 percent failed in 20 years' exposure.

An investigation of galvanized sheets also has been made by Committee A-5. The procedure has been similar to that with iron and steel, except for the definition of a failure, which has been determined by the appearance of base-metal oxide. Data on sheets exposed at Pittsburgh, Altoona, Sandy Hook, State College, and Key West have been reported (14)(15). These show that to date the average life - the time of exposure before base metal oxide appeared - of the lighter coatings was as follows: Altoona, 2.1 years; Pittsburgh, 2.5 years; Sandy Hook, 5.5 years; and State College and Key West, over 8.5 years, the final figure not at this date having been determined.

An investigation, also by Committee A-5, is in progress on hardware, structural shapes, tubular goods, etc., with various metallic coatings. These materials are exposed in the open air on racks. Data from periodical inspections are being reported covering the entire range of deterioration up to destruction of the entire coating. Reports on this work (16) indicate that in general the life of most of the coatings is much longer in rural and sea-coast atmospheres than in those of industrial communities.

Also in progress is an investigation by Committee B-3 by field test methods of corrosion of nonferrous metals and alloys at various locations. Corrosion effects are being determined by change in weight, tension tests, and visual inspection. Reports to date (17) indicate that the loss in tensile strength will be greater in industrial than in rural atmospheres.

Committee D-14 has conducted experiments with nonferrous screen-wire cloth, which was exposed outdoors on vertical frames. Periodical inspections were made and failures noted. A failure was considered to have occurred when there was a break in the wire in at least one place as a result of corrosion. Tension tests were made of the wire before and after exposure. Determination of the sulfur dioxide content of the atmosphere over a period of approximately 1 year were made at or near each of the exposure stations. Quick (18), in reporting the data from this work, has tabulated the condition of the test

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screens at Pittsburgh, Pa., Portsmouth, Va., and Cristobal, Canal Zone, after approximately 9 years' exposure; 19 out of 21 screens had failed from corrosion in Pittsburgh, 3 out of 16 in Portsmouth, and 2 out of 14 in Cristobal. None of similar test screens had failed in Washington, D. C., during the 9 years' exposure. The data may also be expressed as in table 1.

TABLE 1.

Screen material No.	Average period before failure, years (approx.)			
	Pittsburgh	Portsmouth	Cristobal	Washington
1	7-1/2	Over 8-1/2	Over 9	Over 9
2	6-1/2	4	5	Over 9
3	7-1/2	Over 8-1/2	Over 9	Over 9
4	4-1/2	Over 8-1/2	Over 9	Over 9
5	Over 8-1/2 1/	Over 8-1/2	Over 9	Over 9
6	7-1/2	Over 8-1/2	Over 9	Over 9
7	5-1/2	Over 8-1/2	Over 9	Over 9

1/ Final figure not yet determined.

Quick also tabulated the percentage loss of tensile strength after approximately 9 years' exposure. The average loss for all test screens at Pittsburgh was 81 percent, at Portsmouth 53 percent, at Cristobal 51 percent, and at Washington 24 percent. He points out that the deterioration could not be correlated with the results of the determinations of the sulfur dioxide content of the atmosphere at the four locations. Averages of the sulfur dioxide concentrations reported in a previous paper by Quick (19) are 0.19 p. p. m. at Washington, 0.16 p. p. m. at Pittsburgh, 0.26 p. p. m. at Cristobal, and 0.97 p. p. m. at Portsmouth.

Cohen and Ruston (20) have reported data from experiments made with steel rails in England. Eighty-six-pound rails were placed by the side of a railroad line in various locations and weighed at intervals for 8 to 17 years. The annual loss of weight per yard from corrosion was 1.04 pounds in the center of town, 1.48 pounds in a dry section of a smoky tunnel, 1.71 pounds in a wet section of the same tunnel, and 0.18 pound on the seacoast among sand hills. The conclusion from this work was that the amount of corrosion due to smoke in town air appeared to be more than six times that from the atmosphere of the seacoast.

The Joint Corrosion Committee of the Iron and Steel Institute and the British Iron and Steel Federation has computed the probable lives of 2 ounce per square foot zinc coatings on steel in various localities. These are shown (21) to be approximately as in table 2. Since the estimates were based on observations over the period of only 1 year, they are necessarily very approximate.

Richards (22) exposed Nos. 8 and 14 S.W.G. galvanized iron and steel wires to industrial, urban, and rural atmospheres for 5 years, and determined the loss in weight and tensile strength due to rusting. He also obtained data on the loss in weight of ungalvanized mild-steel wire after 2-1/2 years of

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exposure to the same atmospheres. The loss of the galvanizing material due to corrosion in urban and industrial areas was found to be, respectively, about 2.8 and 5.3 times that in rural and suburban areas. The rusting of the ungalvanized-steel wires was about 40 percent greater in urban and 3 times greater in industrial atmospheres than in the pure air of the rural communities. Losses in tensile strength of the galvanized wires were 25 to 50 percent greater in industrial areas than in urban centers, the higher difference being with the lighter wire.

TABLE 2. - Estimated life of a galvanized coating of 2 oz. per sq. ft. of surface when exposed to atmospheric corrosion, years

Type of atmosphere	Locality	Approximate life, years
Overseas	Khartoum	150
	Basrah	80
	Apapa (Nigeria)	70
	Singapore	70
	South Africa	20
Rural	Cardington	25
	Llanwrtyd Wells	25
Suburban	Bournville	15
	Wakefield	10
Urban and industrial	Woolwich	20
	Motherwell	15
	Birmingham	8
	Sheffield	5
Marine	Calshot	25
	Southport	15
Railway tunnel	Dove Holes Tunnel	1

Calculated values for the expected life of No. 8 S.W.G. wire with 0.5 and 1.2 ounces per square foot of galvanizing, based upon the data from these exposure tests and allowing a 25-percent loss in tensile strength, are given by Richards as follows:

	Years
Industrial areas	3.0 and 4.9
Urban areas	5.7 and 9.4
Rural and suburban areas	11.3 and 21.8

Paint

Peters (23) used sulfur dioxide in seeking a suitable accelerated test for paints and found that with high relative humidity the destruction of a paint film was less rapid when the sulfur dioxide concentration was reduced. He painted 5- by 10-cm. iron plates, which had been sand-blasted, and placed them on racks under bell jars containing water solutions of sulfur dioxide. It was estimated that, based upon the volume of the jars, these solutions contained enough sulfur dioxide to give concentrations of approximately

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25,000, 10,000, and 2,500 p. p. m. The jars were removed daily and cleaned, and the solution was renewed. The records of the amount of rusting showed that the higher concentrations attacked the paints so rapidly that small differences in the various paints used could not be detected but that the slower attack of the 2,500-p. p. m. concentration was more suitable. Experiments with 250-p. p. m. sulfur dioxide were so slow in action that this concentration was not useful for an accelerated test.

Leather

Frey (24) investigated the effect of atmospheric exposure on vegetable tanned leather. He concluded that the sulfur dioxide from the air is one of the factors in decay of leather book bindings. Leather binding materials exposed to air and sunlight in Washington, D. C., showed losses as high as 94 percent in tensile strength and tear resistance after a period of 8-1/4 years, while the same materials with very little air and light exposure had essentially their original strength.

Paper

Kimberly (25) investigated the effect of 2 to 9 p. p. m. sulfur dioxide in air on commercial book and writing papers. He hung 8- by 10-1/2-inch sheets of paper in a double-walled sheet-metal and glass cabinet, automatically controlling the temperature at 86° F. and maintaining 65 percent relative humidity. He found that 10 days of such exposure resulted in embrittlement of the papers, as evidenced by decrease in folding endurance, usually of considerable magnitude. Untreated specimens of each paper were tested simultaneously with the treated specimens so that the results might be strictly comparable.

Cloth

Wilkie (26) concluded that sulfur dioxide in the atmosphere is a contributing factor in the so-called "winter damage" to laundered cotton goods. This damage consists of weakening of the fabric so that it tears easily and, in the most advanced stages, crumbles. At times discoloration accompanies the deterioration. He showed, by producing winter damage under controlled conditions, that the presence of a catalyst such as iron, or oxidizing agents left in the cloth from the laundry processes, were necessary for this damage to be severe.

Morgan and Kenalty (27), believing that Wilkie's artificial production of laundry winter damage in the laboratory (26) did not closely approach conditions in actual laundry practice, conducted further laboratory experiments to determine the effect of sulfur dioxide on damp laundered goods. These experiments did not show any clear evidence of deterioration from sulfur dioxide.

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